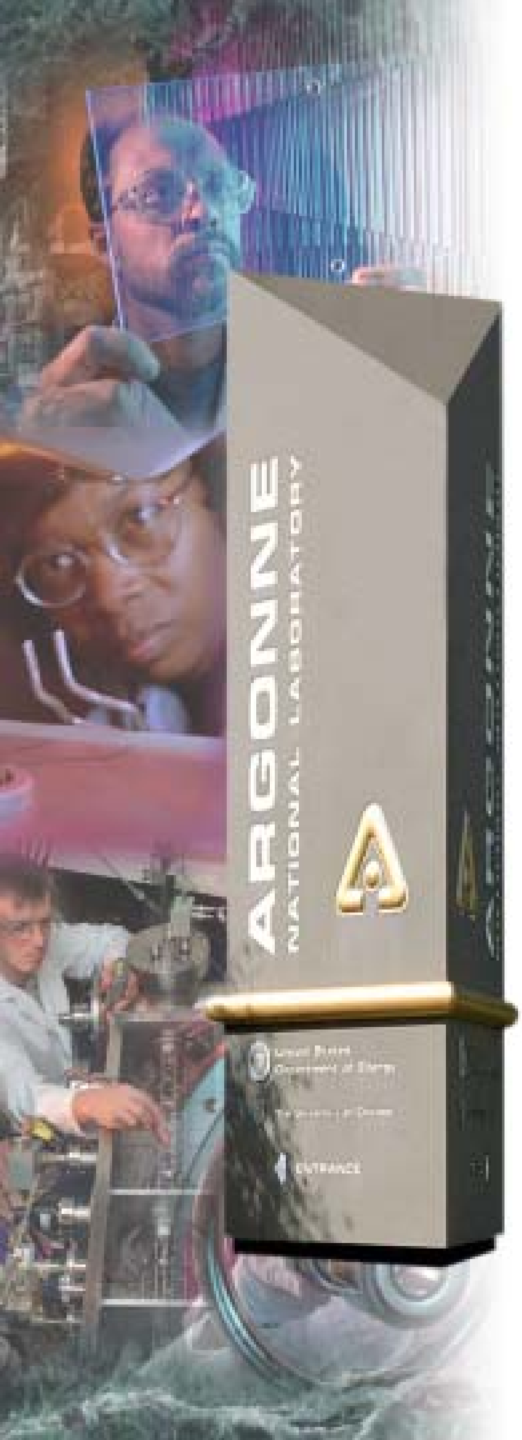


Pump-Probe Studies of Atomic Inner-Shell Photoionization and Vacancy Decay

Steve Southworth

Workshop on Time Domain Science Using X-Ray Techniques
Aug 29 - Sep 1, 2004

Argonne National Laboratory

Office of Science
U.S. Department of Energy

A U.S. Department of Energy
Office of Science Laboratory
Operated by The University of Chicago

Laser-pump X-ray-probe studies of atomic inner-shell physics

- Scientific motivations
- First results at APS beamline 7ID
- Instrumentation and methods
- Future scientific directions
- Suggestions for advanced capabilities at the APS

Collaborators

Atomic Physics Group

Bob Dunford, Dave Ederer, Elliot Kanter, Bertold Krässig, Steve Southworth, Linda Young (ANL-CHM)
Juana Rudati (ANL-APS)

X-ray Optics

Eric Dufresne (ANL-APS)
Peter Eng (Geo-CARS, U. Chicago)

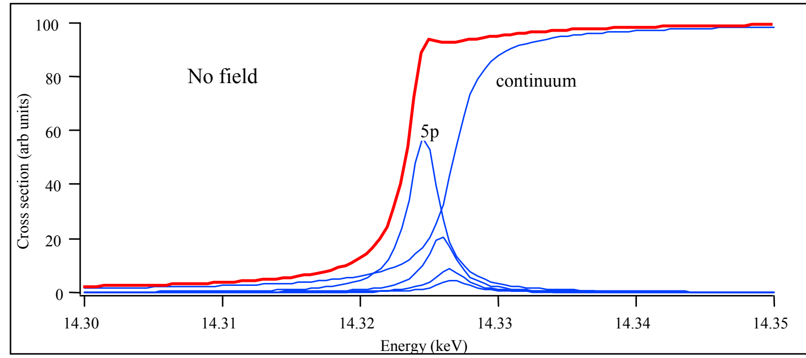
Ultrafast Laser

David Reis, Matt DeCamp, Emily Peterson (U Michigan)
Eric Landahl (ANL-APS)
Rob Crowell, Dave Gosztola (ANL-CHM)

Predicted high-field modification of Kr x-ray absorption edge

Field-free

Kr 1s $\rightarrow$ 5p, 6p, ..., εp

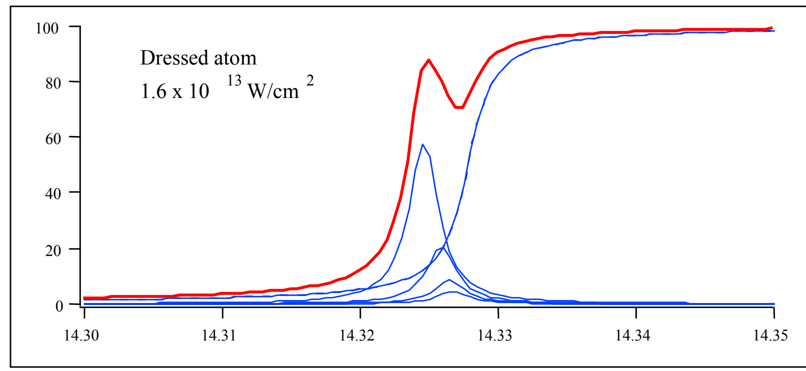


Ponderomotive shift of continuum

$U_p = e^2E^2/4m\omega^2$

$\approx$1 eV for 800 nm laser

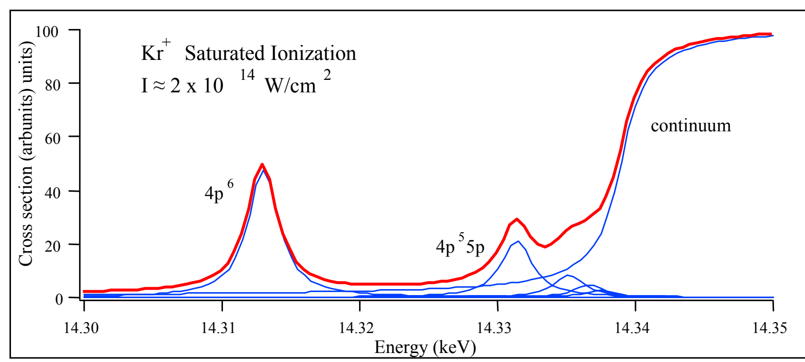
at 1.6 $\times$ 10^{13} W/cm^2



Strong-field ionization

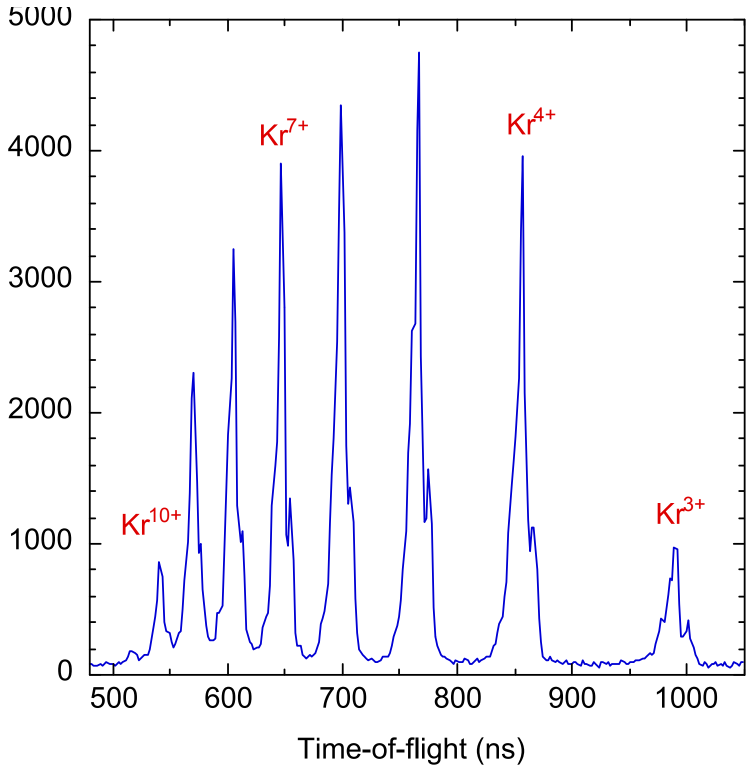
$\approx$2 $\times$ 10^{14} W/cm^2

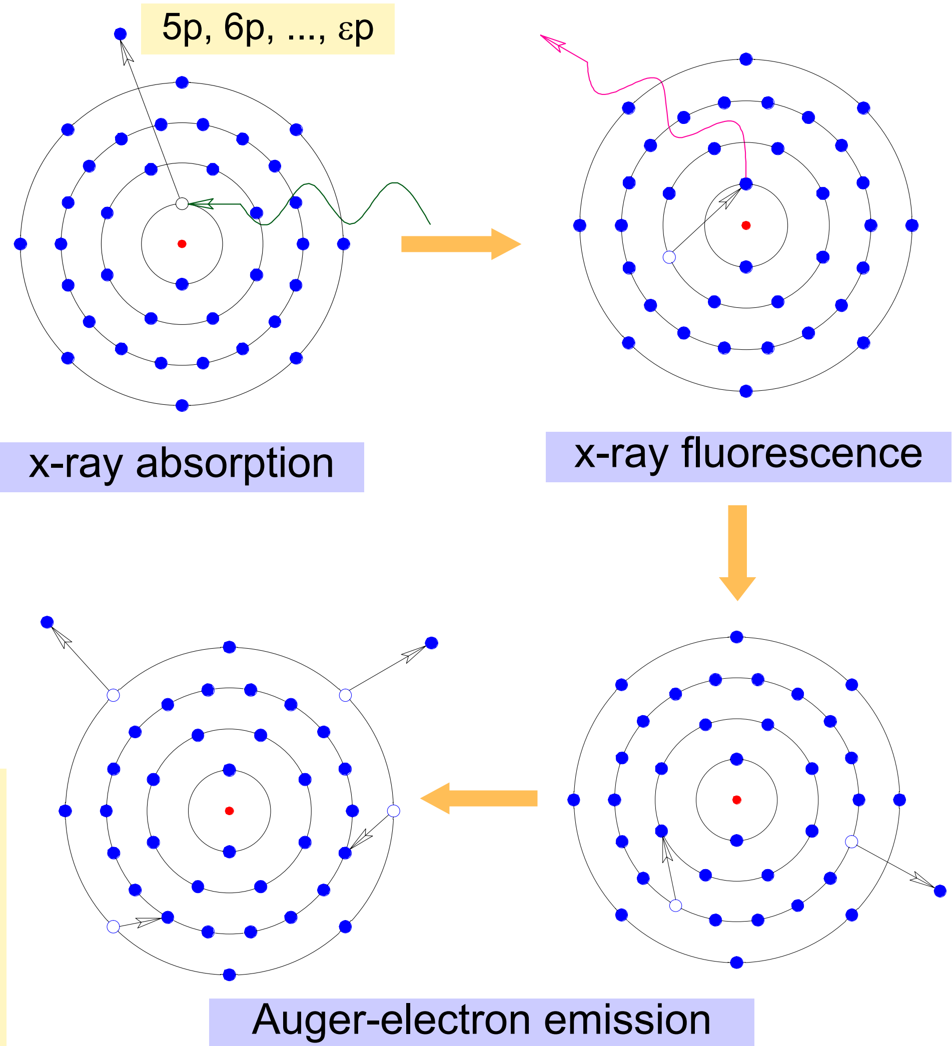
Kr^+ 1s $\rightarrow$ 4p, 5p, 6p, ..., εp



G. B. Armen *et al.*, Phys. Rev. A **67**, 042718 (2003); **69**, 062710 (2004)

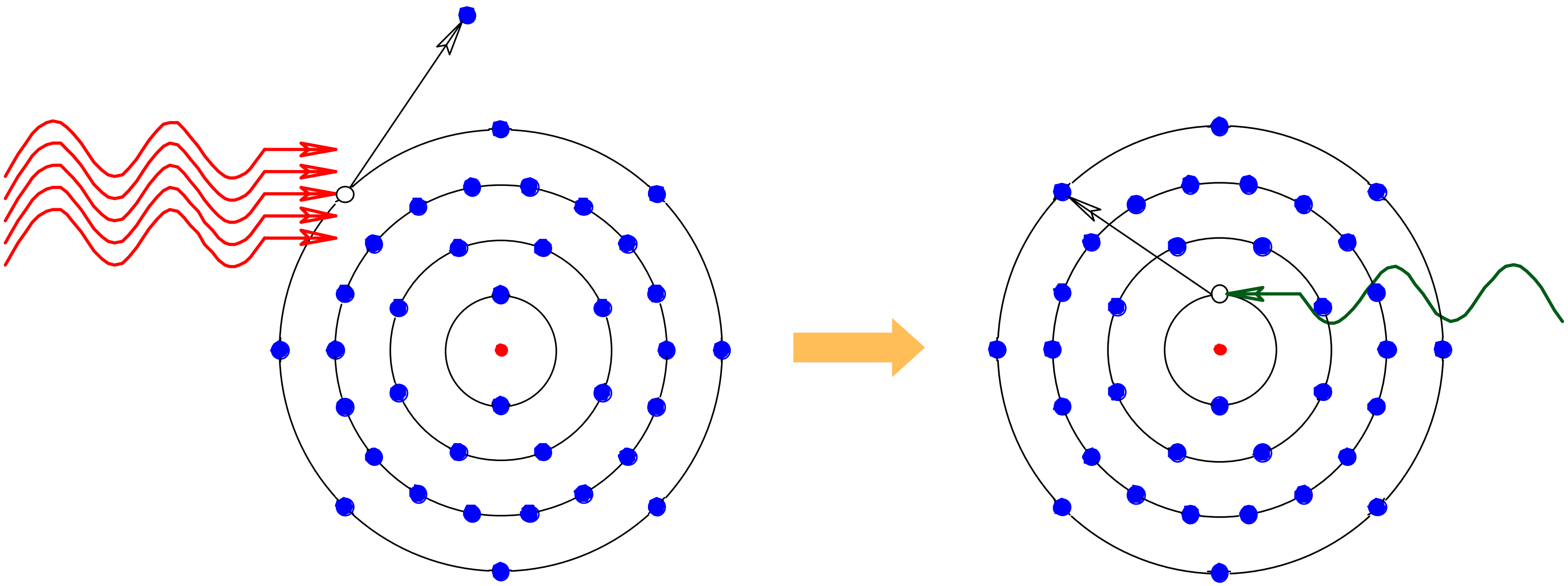
Photoionization and vacancy decay across threshold





- Multi-step vacancy-decay pathways
- Bound and continuum intermediate states
- Post-collision interactions
- Final-state sticking probabilities
- Unknown dressed-atom effects

Our first pump-probe experiment at APS beamline 7ID



High field (≈10^{14} W/cm^2) of optical laser (1.55 eV) ionizes Kr 4p electron

X ray (14.3 keV) excites 1s electron to 4p hole

D. A. Reis, P. H. Bucksbaum, M. F. DeCamp, Rad. Phys. Chem. **70**, 605 (2004)

Beamline 7ID laser system - June 2004

1.1 km APS storage ring

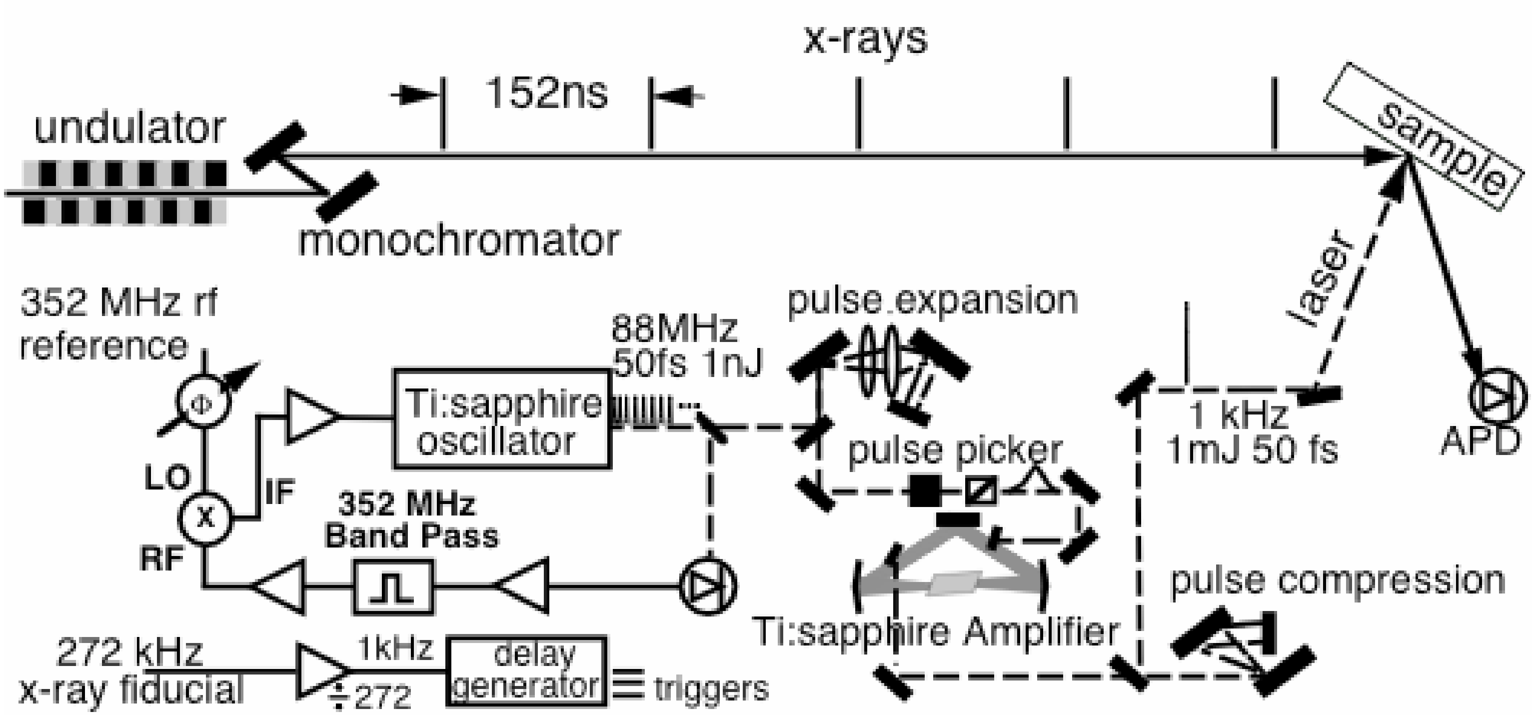
352 MHz RF, 1296 buckets

272 kHz, 3.68 μs

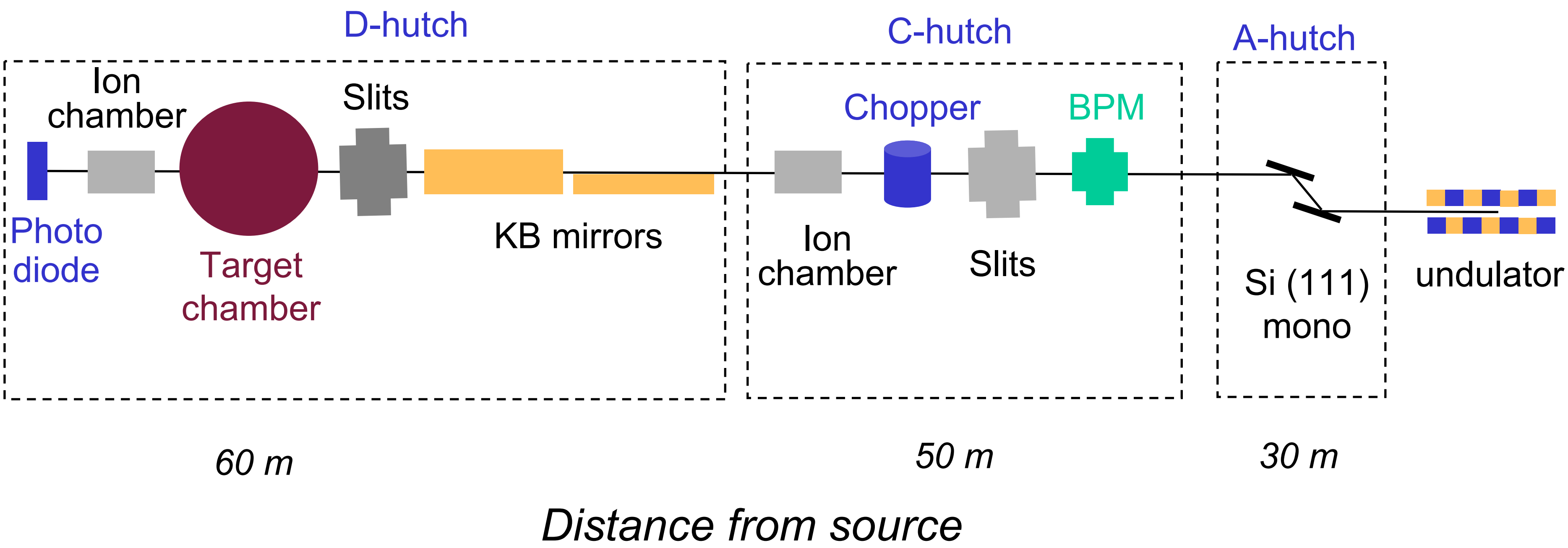
laser-xray timing jitter ≈2 ps

oscillator: 88 MHz, 1 nJ, 50 fs
amplifier: 1 kHz, 1 mJ, 50 fs

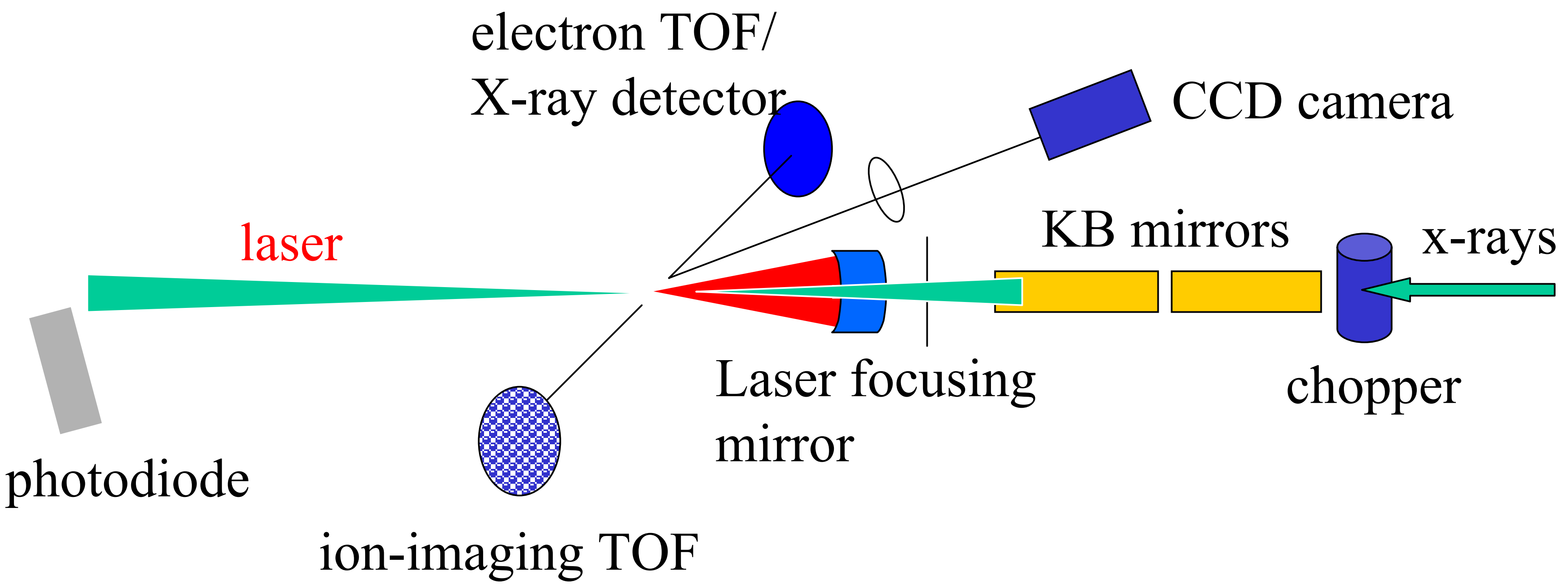
focusing to ≈10 μm spot required for 10^{13}–10^{14} W/cm^2 regime



7ID x-ray beam path

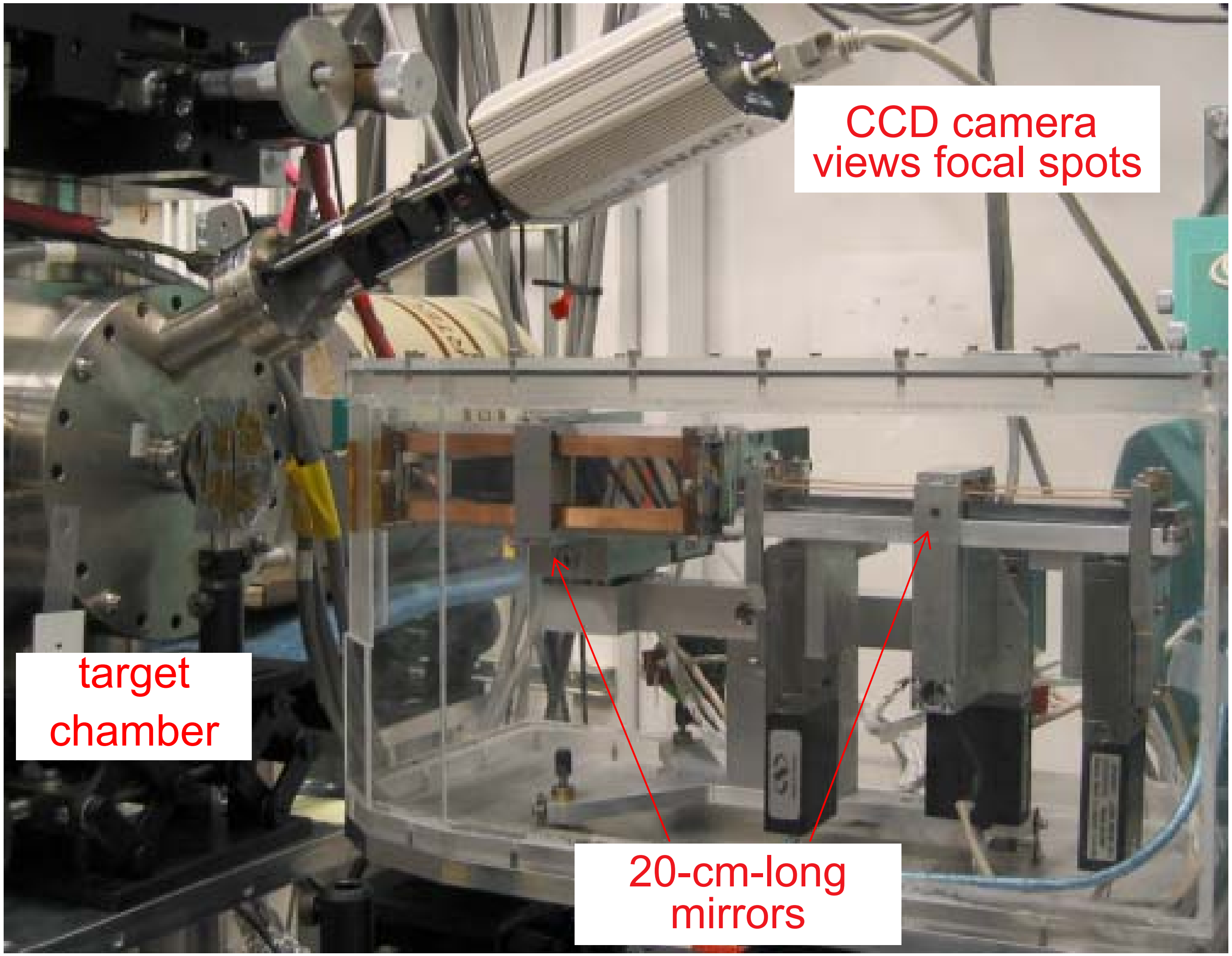


Focused laser and x-ray beams overlapped in space and time through atomic-gas target



P.J. Eng, M. Newville, M. L. Rivers, S.R. Sutton, SPIE Vol. 3449, 145 (1998)

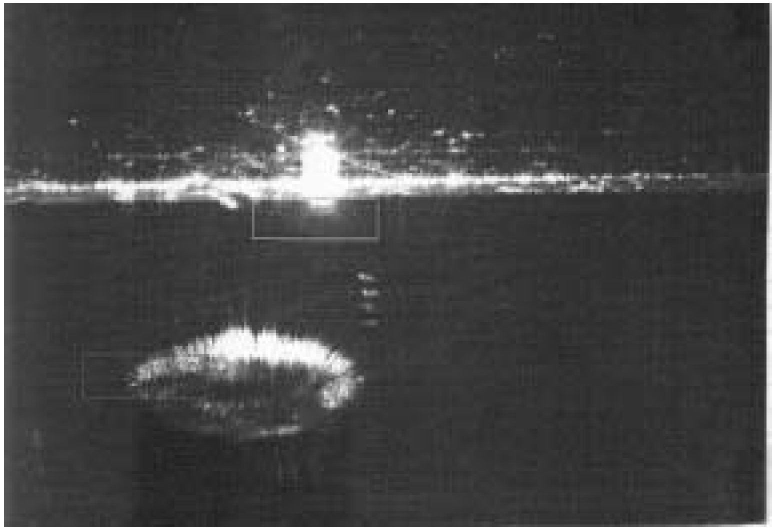
Kirkpatrick-Baez x-ray microfocus mirrors



Courtesy of Peter Eng, GeoCARS, U. Chicago

Viewing and overlapping focal spots

- Focus x rays to center of chamber
- Locate x-ray centroid with BGO
- Overlap focused laser
  - -rough: BGO crystal
  - -fine: in-vacuum cross hairs



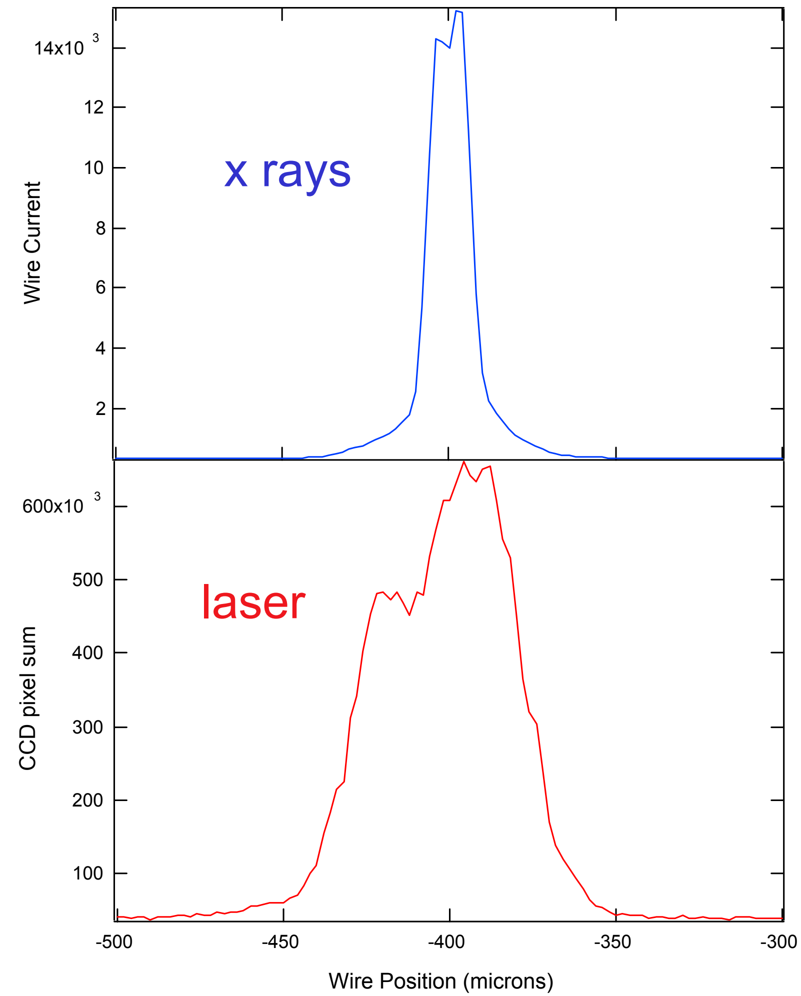
Measuring overlap of focused laser and x-ray beams

Scan 10 μm cross-hair

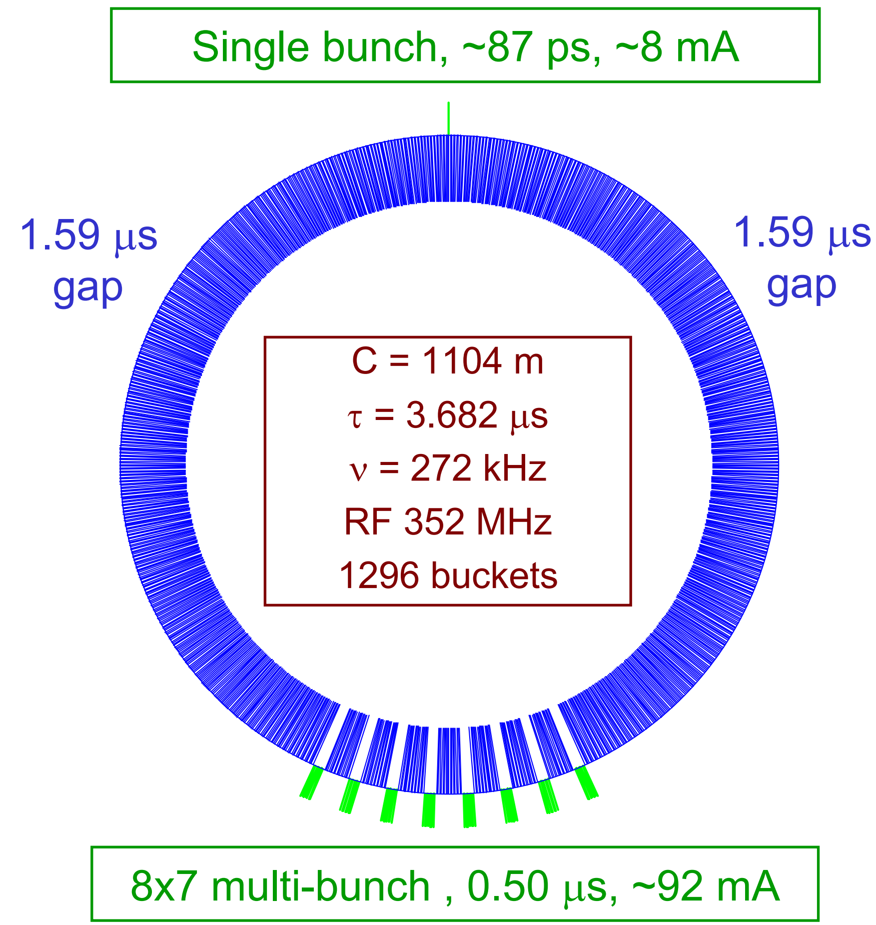
X-rays: monitor current
electrically isolated cross-hair

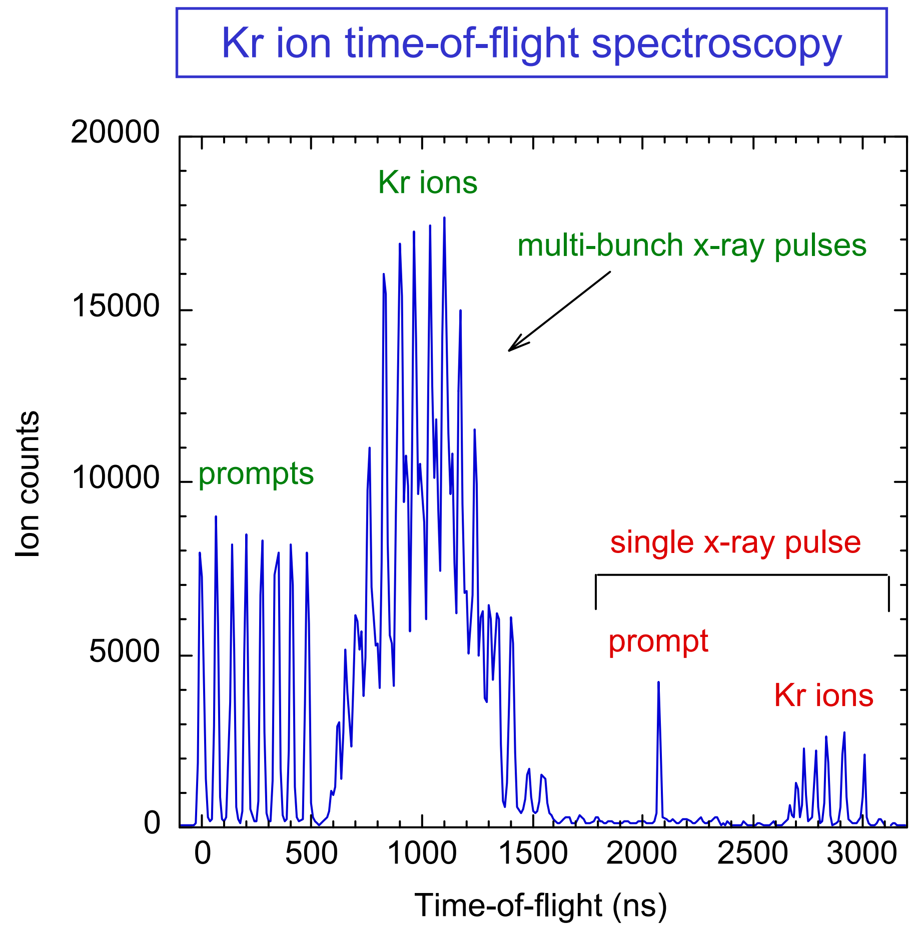
Laser: monitor scattered light
pixel sum from a selected region on CCD camera

June 2004:
Laser waist ≈ 30 μm
Rayleigh range ≈ 3 mm
X-rays ≈ 13 μm
Crossing angle ≈ 20 μm/mm



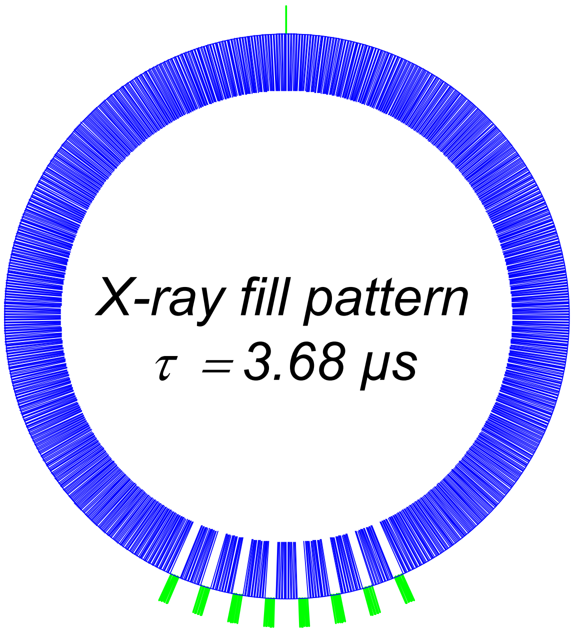
APS ring fill pattern with isolated bucket





A. McPherson, J. Wang, P. L. Lee, D. M. Mills, J. Synch. Rad. **7**, 1 (2000)

Mechanical chopper transmits singlet x-ray pulses



≈80,000 rpm air-bearing rotor

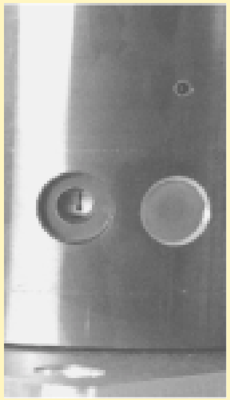
drive frequency phase locked to ring RF

transmits 1 singlet x-ray pulse out of 102

transmitted flux ≈ $(8 \times 10^{-4}) \times$ total flux

KB-focused flux ≈ 4×10^{5} x-rays/pulse

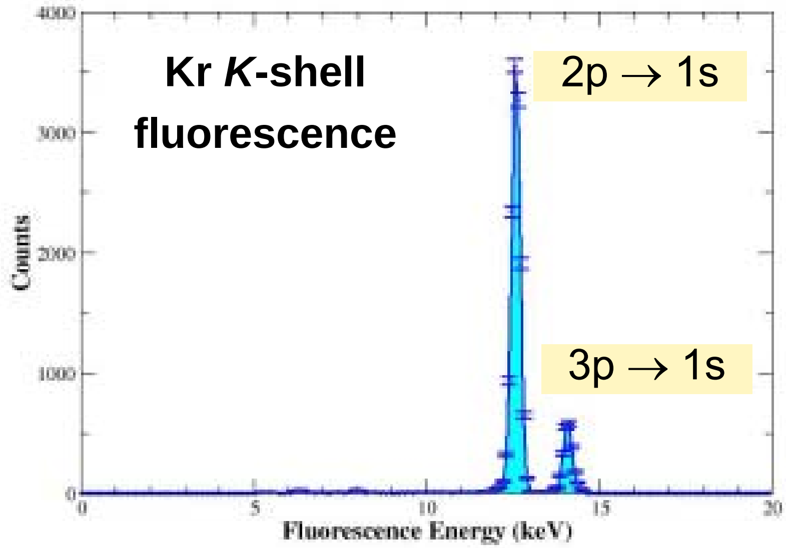
Chopper selects singlet x-ray pulses @ 2.66 kHz
Laser @ 887 Hz : 1 laser-on vs 2 laser-off

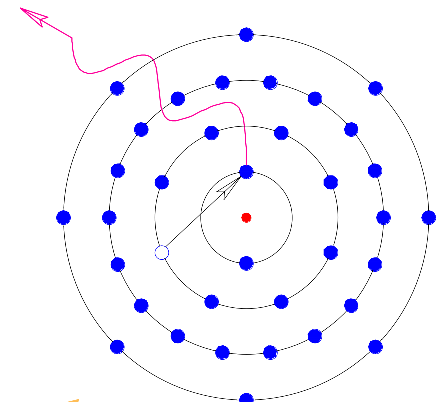


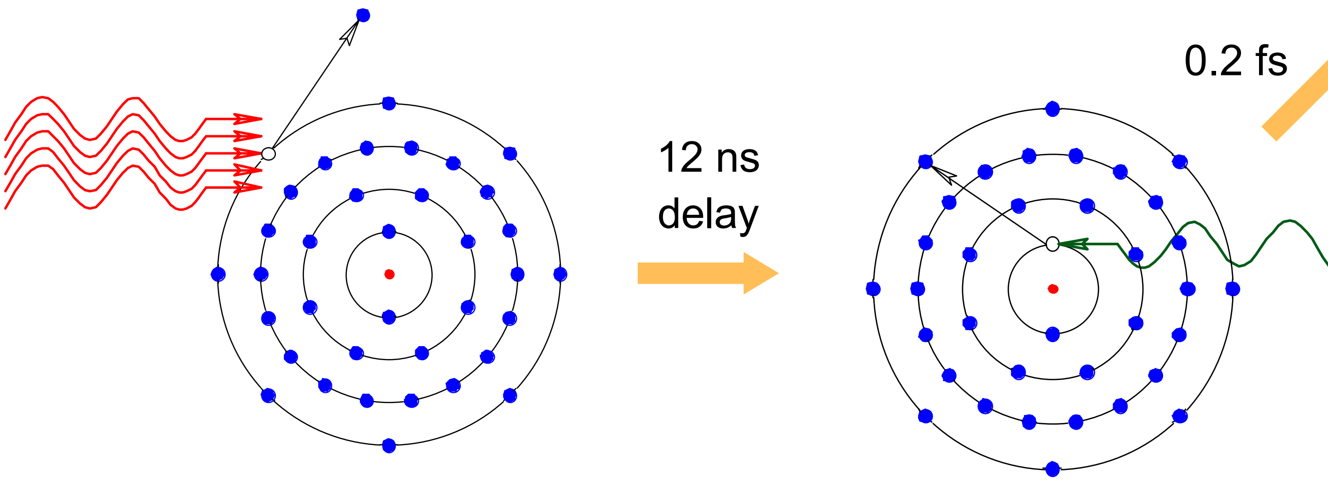
0.51 mm slot
50.8 mm diameter
2.45 µs open time

Laser: 800 nm, 887 Hz, 0.3 mJ, 60 fs, 30 μm focus, 6×10^{14} W/cm^2

X-ray probe of laser-produced Kr^+

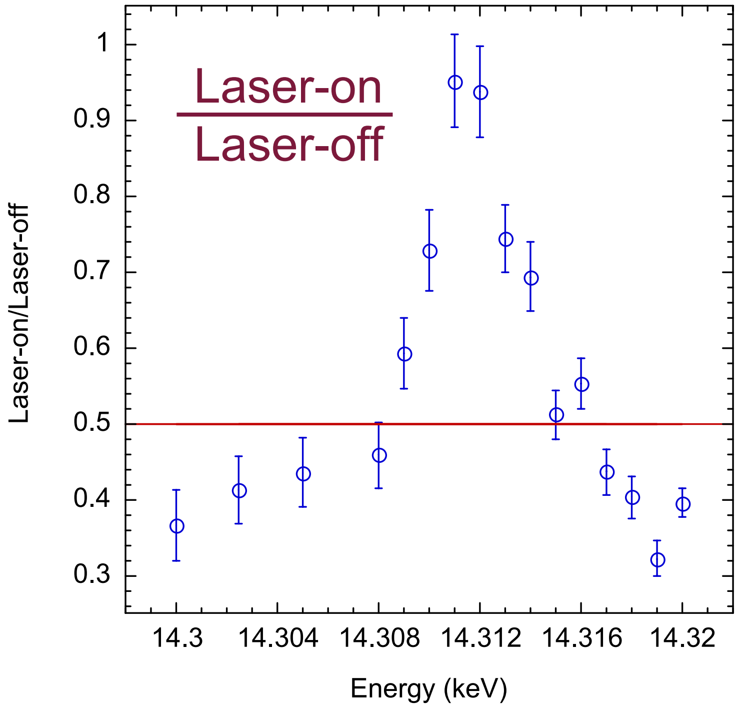






1s → 4p resonance in laser-produced Kr^+

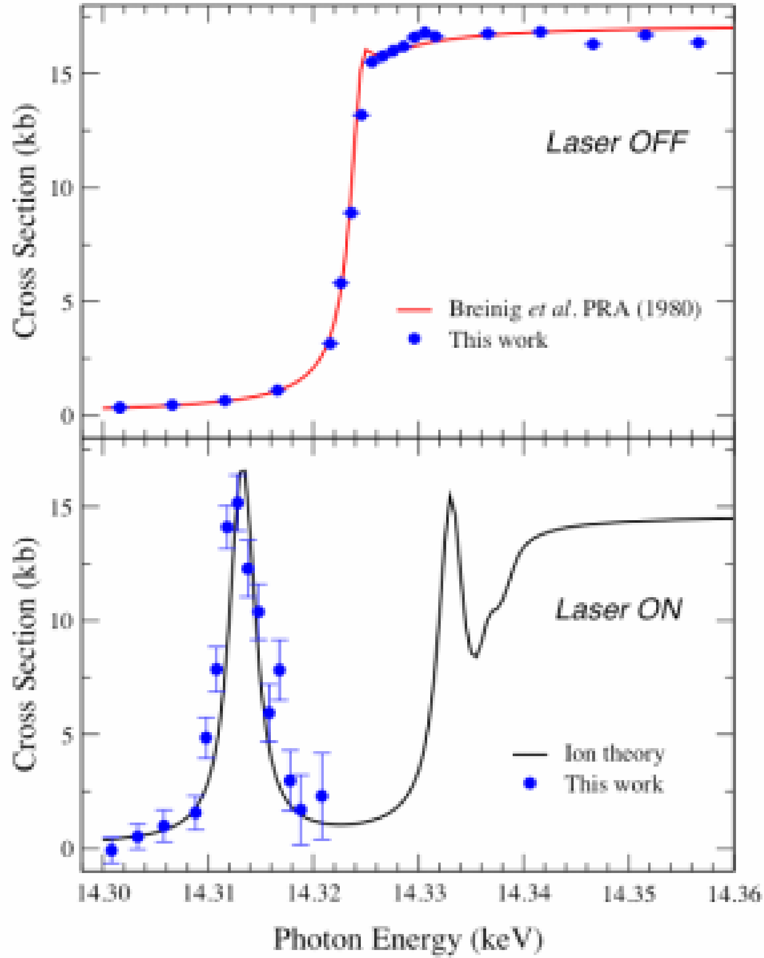
x-ray fluorescence rate



887 Hz laser pulses

2661 Hz x-ray pulses

on/off = 0.5



Kr^+ theory: L. Pan and D.R. Beck

Coulomb explosion of Kr^+ ion assembly

focal volume ≈30 μm dia × 3 mm long

intensity ≈6 × 10^{14} W/cm²

atom density ≈10^{13}/cm³

→10^{7} ions/pulse

10 eV Kr^+ ion velocity ≈ 40 μm/ns

→ study dynamics of ion assembly

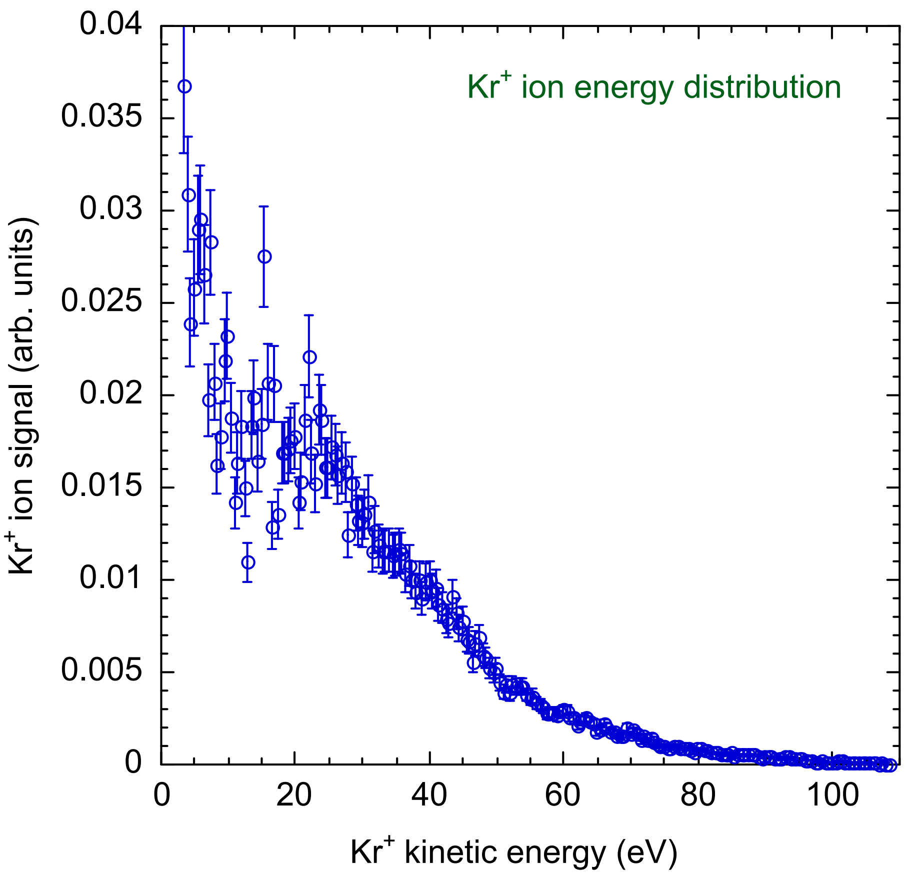
by varying x-ray probe delay

similar to Coulomb explosion of laser-ionized clusters

tunable x-ray probe

≈10 μm spatial resolution

≈100 ps temporal resolution

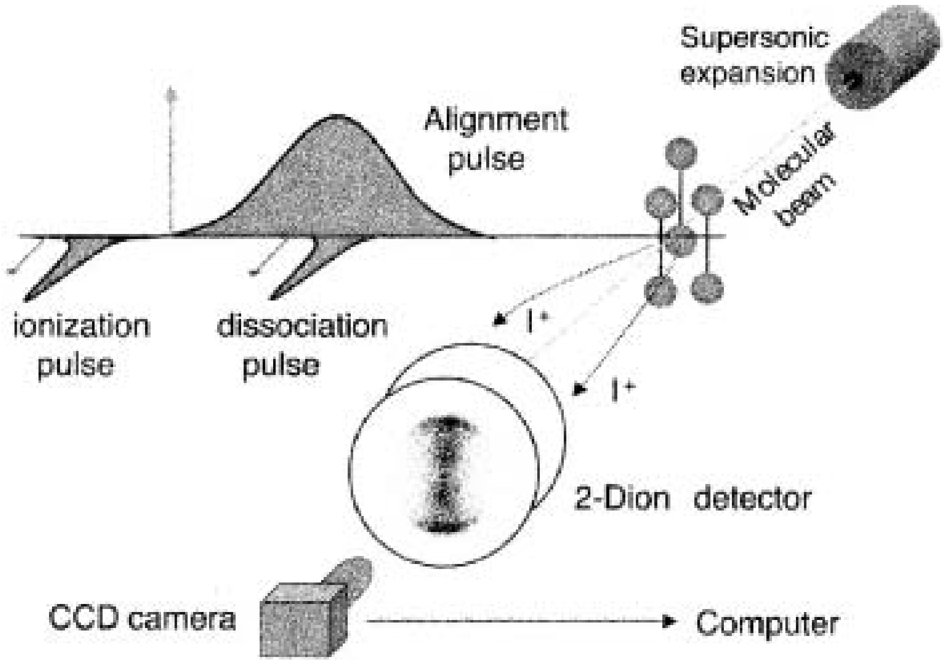


Laser upgrades in progress at beamline 7ID

- Dedicated laser hutch
- Ti:sapphire regenerative amplifier
- Diode-pumped solid-state pump laser
- Compressed pulses 2.5 mJ, 40 fs – 10 ps
- Stretched pulses 4.0 mJ, 130 ps
- Repetition rate 1 – 5 kHz

→ Higher peak intensity, higher rep rate, and complete overlap of x-ray pulses (87 ps) for dressed-atom experiments

Future directions: atoms → small molecules



Alignment, molecular geometry, coherent control

Marcos Dantus

Tamar Seideman

Steve Pratt

Stefan Vajda

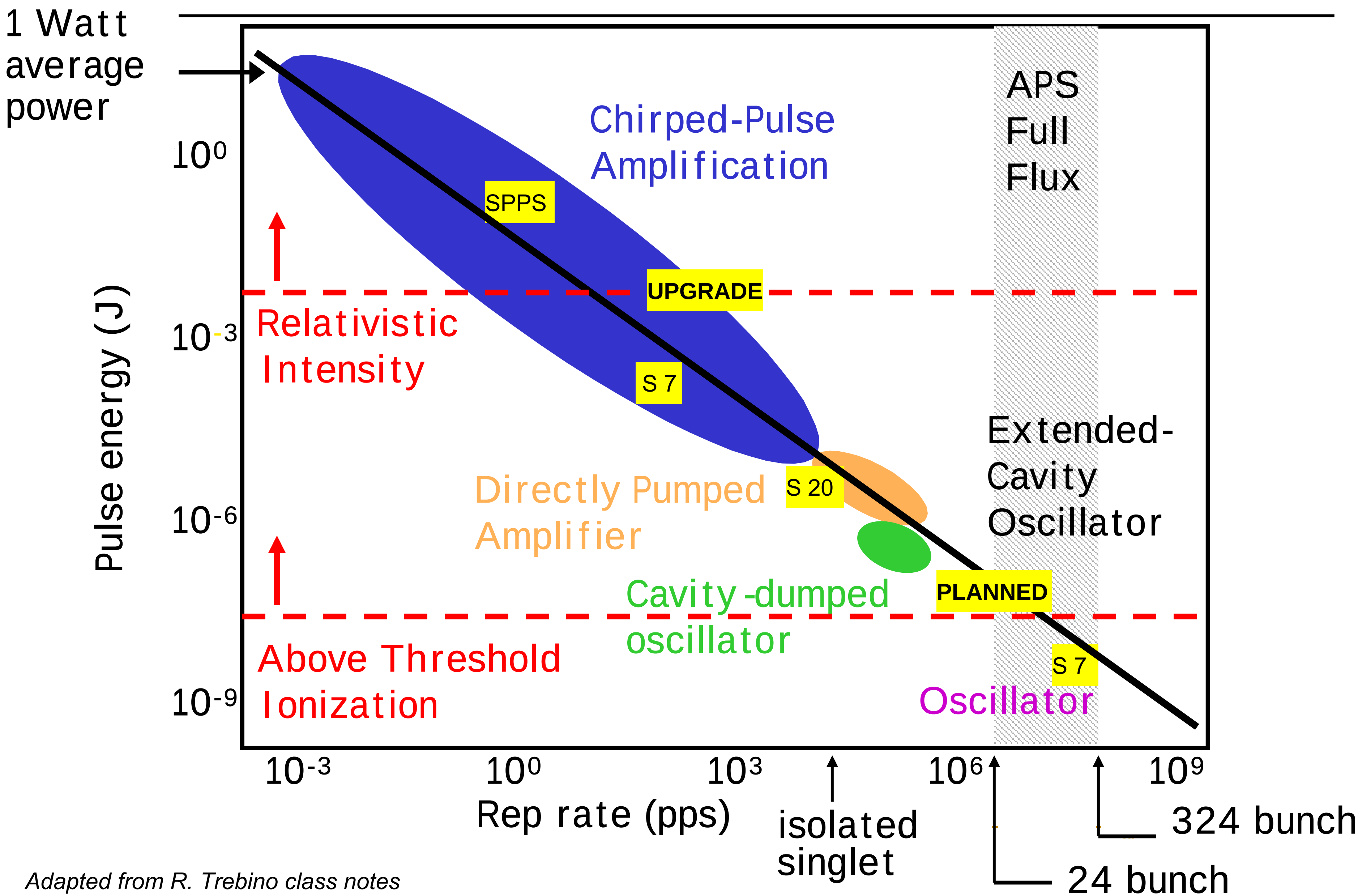
Aligning molecules with laser pulses

H. Stapelfeldt and T. Seideman

Rev. Mod. Phys. **75**, 543 (2003)

courtesy of E. C. Landahl

Femtosecond-laser pulse energy vs. repetition rate



Adapted from R. Trebino class notes

Considerations for improved pump-probe capabilities

Repetition rate

Storage-ring bunch patterns
Mechanical or x-ray-optical choppers
High-power laser oscillators

X-rays/pulse

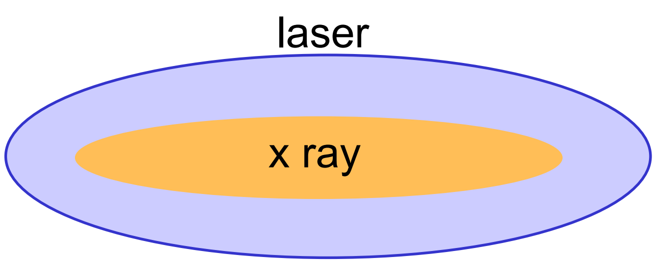
high stored current in singlets
pink-beam experiments

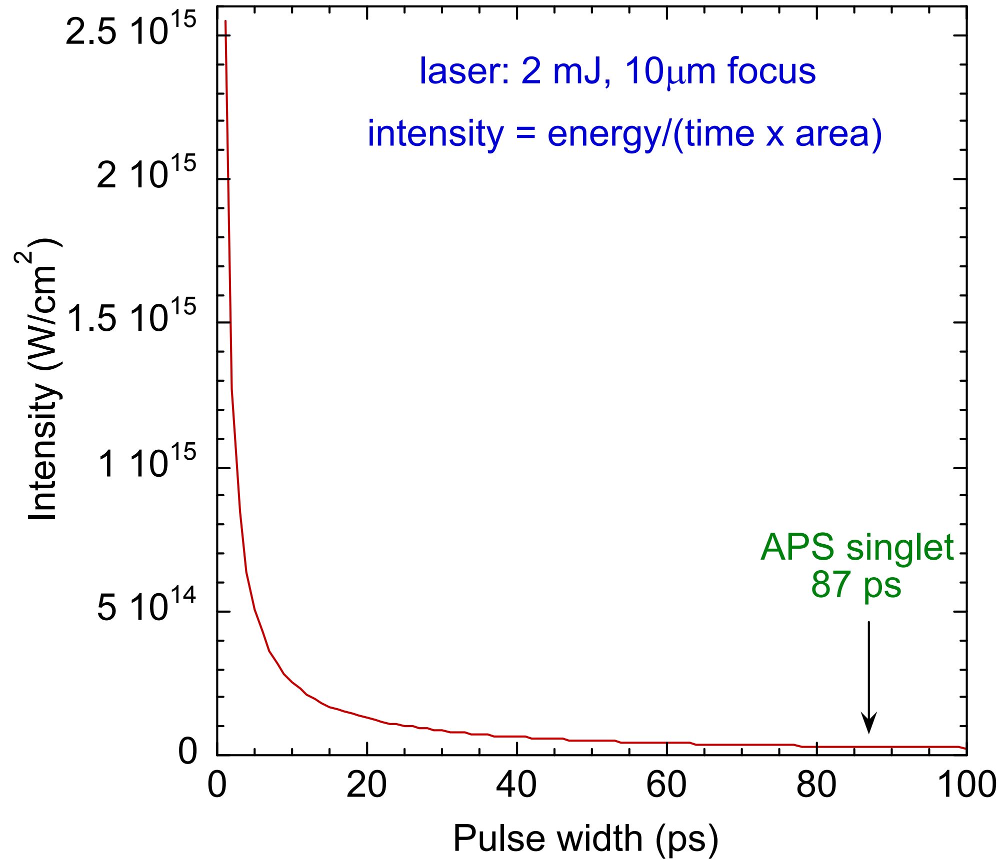
X-ray focusing with high throughput

KB mirrors
Li lens
zone plates

APS x-ray pulse width

Shorter x-ray pulse enables dressed-atom experiments at higher laser intensity





Develop APS capability for pump-probe experiments using soft x rays

- Lower-Z atoms at lower x-ray energies
- Longer lifetimes → better resolved resonance and threshold structure
- Fewer electrons → simpler decay spectra
- Larger cross sections
- Can do high-resolution electron spectroscopy, but x-ray spectroscopy more challenging